

## ULTRASONIC ATTENUATION IN GELS

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**ABSTRACT.** In this paper the ultrasonic propagation in gels is studied from the viewpoint of attenuation of the incident waves. In a theoretical discussion the propagation in a medium having spherical particles suspended in a liquid medium has been evaluated. The paper shows that the attenuation per unit volume of such a medium is given by,

$$\alpha = 2\pi n a^3 (\rho_0/\rho') q \cdot k.$$

if the effect of scattering is to be considered too. The theory points out the different cases showing the relative importance of these terms. Considering the Lamb and Sewell theory in details, a starting point for the present author's results is also indicated

From the theoretical considerations given in this paper, the attenuation is calculated for a few gels also showing its dependence upon temperature. Furthermore, the results are satisfactorily explained by the theory outlined in the paper.

Using an ultrasonic pulse technique, the attenuation for a few gels has been practically determined and its variation with temperature and frequency studied. The diminution of the kinematic viscosity at higher temperatures and the increase in the scattering account for the values of attenuation showing a maximum near about 55°C.

## INTRODUCTION

The propagation of sound waves in a medium containing suspended particles was first studied by Lord Rayleigh (1896). Lamb (1884) took up the same problem in a more general way, considering the various modes of vibration of a sphere that scattered the incident wave. Herzfeld (1930) developed a theory for sound propagation in suspensions by extending the Rayleigh boundary conditions. Sewell (1911) studied the problem as applicable to such natural suspensions as fog and smoke particles in air. He calculated the loss of energy in case of spherical particles.

Sound propagation in aerosols has recently been studied by Brandt (1937), Heidmann, Freund and Brandt (1937). Further, the acoustic impedance of such a foggy atmosphere was investigated by Ghosh (1936) who tabulated a large amount of significant data based upon his theoretical results for particles of radius ten microns and lesser. Richardson (1938) and Epstein (1942); Hartmann and Focke (1940) developed an ultrasonic technique using a piezo-electric oscillator to study the absorption of sound in artificial fogs and smoke. Richardson's results were the first step towards the practical verification of the Lamb and Sewell's theory. Following this, Urick (1948) applied an ultrasonic pulse technique to obtain the attenuation of sound

waves in artificial smokes and also in sand and kaolin particles suspended in water. These results also supported the Lamb and Sewell theory.

It is evident, therefore, that no significant data exists regarding the factors affecting the propagation of sound waves in a gelatinous medium. The composite nature of these gels must necessarily affect the attenuation in such a medium, where a solid phase enmeshes and entrains an overwhelmingly larger proportion of the fluid which is usually water.

#### THEORETICAL CONSIDERATIONS

Transmission of sound in fog and smoke has been studied by Lamb and Sewell. Owing to its great inertia in comparison with that of an equal amount of air, a globule of water in suspension, if not too small, may remain practically at rest as the air waves beat upon it. If, however, the radius be diminished, the inertia diminishes as  $a^3$ , and the surface on which viscosity acts diminishes as  $a^2$  and it is expected that a stage will be reached when the globule will simply drift to and fro with the vibrating air, and so causes little or no loss of energy.

#### *Scattering of Sound in Suspensions*

The scattering of waves in suspensions can be divided into two classes :

- (a) The Rayleigh scattering and,
- (b) the Lamb and Sewell scattering.

In the former case it is well established that scattering increases with the decrease of wavelength or the particle size. For larger wavelengths  $ka \ll 1$ , and the amplitude of the particles in vibration is given by,

$$1 - \frac{\rho_1 - \rho_0}{\rho_1 + \frac{1}{2}\rho_0}$$

where  $\rho_1, \rho_0$  are the densities of the solid and the fluid respectively,  $a$  is the radius of the particles and  $k$  is the propagation constant ( $2\pi/\lambda$ ), in which  $\lambda$  is the wavelength.

In the second case, the viscosity is taken into consideration, when it is seen that the scattering gives rise to additional terms in the velocity potential of the scattered waves ; the particles are dragged to and fro with the waves and the resulting amplitude is given by,

$$1 + \frac{\sigma \xi}{k} = \frac{\rho_1 - \rho_0}{\rho_1 - \rho_0 + 3\rho_0 a_1} \quad \dots (2)$$

$$\text{where,} \quad a_1 = \frac{1}{2} \left( 1 + \frac{3}{2} \frac{1}{a\beta} \right) - \frac{3}{2} \frac{j}{a\beta} \left( 1 + \frac{1}{a\beta} \right) \text{ for } a\beta < 1 \quad \dots (2)$$

and,  $\beta = \sqrt{\frac{\sigma}{2\nu}}$ ;  $\sigma$  is the angular frequency given by  $2\pi f$ , and  $\nu$  is the kinematic

viscosity. The behaviour of the particles depends upon  $\beta$ , i.e., on value of the viscosity. For the particles of larger diameter, the amplitude is the same as that of the oscillating fluid at larger wavelengths. This contention can be upheld considering the following tables which show the values of the amplitude of the particles at different wavelengths for two particle sizes.

TABLE I  
Radius  $a = 10^{-3}$  cm.

$\lambda$	$L$	$\lambda$	$L$
5	.019	100	.347
20	.074	400	.833
40	.147	800	.955
60	.217	Inf.	1.000
80	.284		

TABLE II  
Radius  $a = 10^{-4}$  cm.

$\lambda$	5	10	25	30	100	Inf
$L$	.855	.961	.990	.996	.9996	1.0000

For particles of smaller radii, it follows from the foregoing that the amplitude of the free particles will be practically zero at these smaller wavelengths (corresponding to megacycle frequencies). Table III presents the calculated values of the amplitudes which show that the value is practically zero.

TABLE III  
The amplitude of the particles at different values of the radius for frequency of 10 megacycles per second.

$a$	$\frac{1}{2} \left( 1 + \frac{3}{2a\beta} \right)$	$\frac{1}{2} \frac{1}{a\beta} \left( 1 + \frac{1}{a\beta} \right)$	$a_1$	$\frac{\sigma \xi}{k} + 1$	$\xi$ real
$10^{-4}$	.45	.7155	.45-.72j	1.45-.72j	$34 \times 10^{-6}$
$10^{-5}$	4.91	30.27	4.91-30.27j	5.91-30.27j	$.60 \times 10^{-8}$
$10^{-6}$	44.60	2646.0	44.6-2646.0j	45.6-2646.0j	$.6 \times 10^{-9}$

The case of the suspended fog and smoke particles in a gaseous medium known as aerosols has been investigated by Richardson (1934). Particular cases of smokes of stearic acid, magnesium oxide and lycopodium dust are included in his work. Lamb's scattering formula is used to determine the sound absorption coefficients. Remarkable agreement between the theoretical and practical values is obtained. In a particular case,

Particle size,  $a = 2.5 \cdot 10^{-4}$   
 Viscosity,  $\nu = .15 \text{ cm/sec.}$   
 Number of particles per c.c.,  $n = 1.5 \cdot 10^6$   
 Velocity of the waves  $= 3.4 \times 10^4 \text{ cm/sec.}$   
 Frequency  $= 42 \text{ Kc./sec.}$   
 Attenuation per unit volume  $= .0301$  (theoretical)  
 $= .029$  (practical)

In recent years Urick (*loc. cit*) studied the absorption of sound waves in sand and kaolin particles soaked in water at the megacycle frequencies. His result also shows an agreement with the above theoretical evaluations. Table IV is based on his work.

TABLE IV

The absorption values for kaolin and sand particles.

Frequency Mc/sec.	Kaolin one per cent .9 $\mu$		Fine sand one per cent 2.2 $\mu$	
	Observed	Calculated	Observed	Calculated
1.0	0.024	0.038	0.028	0.032
3.0	0.087	0.018	0.093	0.095
8.9	0.256	0.324	0.232	0.234
15.0	0.450	0.500	0.385	0.347

#### SOUND PROPAGATION IN GELS THE COMPOSITE MEDIUM

According to the present day conceptions, a gel consists of a number of particles *viz.* barium sulphate, iron silicate, thorium phosphate, etc., embedded in a fluid medium. These particles are surrounded by a layer of the fluid which acts in binding one particle with another and provides a structural bond. Thus the contiguous particles form a composite medium with the intervening liquid that is unattached to the particles. In fact, it is this absorbed layer that prevents the formation of coarser aggregation from particles of colloidal and molecular dimensions. Bradford points out that the formation of gel by cooling a sol is a case of crystallisation. Moeller (1921).

is in general agreement with the above views. Weimarn (1908, 1909) concludes from his investigations that a gel consists of highly crystallized granules soaked in the dispersive medium from a supersaturated solution. The process of a sol-gel transformation is a continuous process and there is no evidence on record to show that the aggregation of the particles changes suddenly during the transformation.

Thus Walpole (1913) showed that the refractive index changes slowly but continuously during the gelation of a sol. Ghosh (1930) supports this view and shows, from the measurements of extinction coefficients by the Nutting spectrophotometer in the region of  $6000^{\circ}\text{A}$ , that the changes are continuous and gradual. In a series of papers the elastic constants of certain gels have also been evaluated (Srivastava, 1950). These changes in the values of the Young's modulus and other moduli show a continuous variation and confirm the above views (Srivastava, 1951).

From this experimental support it becomes evident that the gel structure maintains its form during the sol-gel transformation. This gives more reason to the belief of the existence of a double layer as postulated above.

Furthermore, the X-ray work on gels carried on by Debye and Hukel (1924) confirms the above view-points. It is therefore evidently clear that there is a considerable difference between a medium of this type on the one hand and another of the type of sand and kaolin particles in water which has particles entirely independent of one another. In order, therefore, to account for the wave propagation in such a medium we have to take into consideration the density and the bulk modulus of the composite medium. The density of the composite medium will be given by,

$$\rho' = \beta\rho_1 + (1 - \beta)\rho_0 \quad \dots (3)$$

where,

$$\beta = \frac{4\pi na^3}{3} \quad \dots (4)$$

$n$  being the number of particles per c c.,  $\rho_1$  and  $\rho_0$  are the densities of the solid and the liquid phases respectively. The bulk modulus of the medium will be obtained as follows : For an applied pressure  $P$ , the change in volume of the gel will be say  $dV$  in an initial volume  $V$ . Then neglecting all interaction between the solid and the liquid phases, this total change  $dV$  will be made up of the sum of the changes in the volume of the solid and the liquid phases. Thus,  $dV = dV_0 + dV_1$ . Also from the definition of the bulk modulus we have,

$$P = K_0 \frac{dV_0}{V(1 - \beta)} = K_1 \frac{dV_1}{V\beta} \quad \dots (5)$$

where  $K_1$  and  $K_0$  are the bulk modulus of the solid and the liquid phases. The composite bulk modulus is given by,

$$K' = \frac{K_1 K_0}{K_1(1-\beta) + K_0\beta} \quad (6)$$

It is of interest to note here that in case the porosity  $\beta=0$ , the liquid phase only exists and the value of the composite bulk modulus  $K'=K_0$  only. This result shows that the formula derived for  $K'$  is adequate for the extreme case when  $\beta=0$ . Similarly, when  $\beta=1$ , the formula yields  $K'=K_1$ , showing that the bulk modulus is that due to the solid phase alone. This is also clear from the porosity expression because the value  $\beta=1$ , implies that the entire volume of the composite medium is made up of the solid phase. The formula, therefore, is valid for all values of  $\beta$  lying between unity and zero. In practice, however, it will be seen later on that the values of  $\beta$  with which we are concerned in the case of these gelatinous substances lies between 2% to about 10%.

As shown elsewhere (Srivastava, 1949), the formula for the composite bulk modulus, as shown above, gives results of the bulk modulus on assuming the values of  $K_0$  and  $K_1$ , and  $\beta$ , which are in fair agreement with the practically determined values. These results confirm the validity of the above theoretical results and give ground enough to treat the above gel structure established.

#### SOUND PROPAGATION IN GELS WITH SCATTERING

In the treatment so far no mention has been made of the scattering of the waves by the solid particles in a gelatinous medium. The treatment given below includes the scattering effects due to the sphere. We now proceed to evaluate the total effect of all the secondary vibrations that issue forth from the composite medium of thickness  $\Delta x$  which is large compared to the wavelength. In figure 1, the secondary vibrations at a

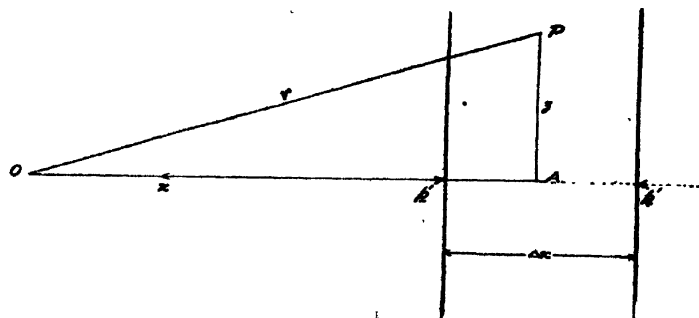


FIG. 1

The forces acting upon the composite element  $\Delta x$

distant point  $O$  due to a particle placed at  $P$  are denoted by the velocity potential

$$\phi' = A_0 f_0(kr) + (H + jK) f_1(kr) \cdot kr \cdot \cos \theta + \dots \quad (14)$$

where  $OP = r$ . If  $AP = z$ , then consider the element of thickness  $\Delta x$  having a volume  $2\pi z dz \Delta x$  which contains  $2\pi z dz \Delta x \cdot n$  particles.

$$\text{Also,} \quad r^2 = x^2 + z^2, \text{ and, } z dz = r dr; \cos \theta \simeq 1 \quad \dots (15)$$

For distant values of  $r$  the resultant of all the secondary vibrations issuing forth from the stratum will be given by,

$$\phi' = 2\pi n \Delta x \int_{-x}^{\infty} \left[ A_0 \frac{e^{-jkr}}{kr} - (H + jK) j \frac{e^{-jkr}}{kr} \right] r dr \quad \dots (16)$$

or,

$$\phi' = 2\pi n \Delta x [A_0 j + H + jK] e^{jkr} \quad \dots (17)$$

The coefficients  $A_0$  and  $(H + jK)$  have been calculated by Lamb. They are,

$$A_0 = -\frac{1}{3} k^3 a^3 \left[ 1 - \frac{k^2 a^2}{2} + j \frac{k^3 a^3}{3} \right] \quad \dots (18)$$

$$H + jK = j a_1 (P + jQ) k^3 a^3 \quad \dots (19)$$

where,

$$a_1 = \frac{1}{2} \left( 1 + \frac{3}{2a\beta} \right) - \frac{3}{4} \frac{j}{a\beta} \left( 1 + \frac{1}{a\beta} \right) \left. \vphantom{\frac{1}{2} \left( 1 + \frac{3}{2a\beta} \right)} \right\} \text{For } a\beta \ll 1 \quad \dots (20)$$

$$P + jQ = (\rho_1 - \rho_0) / ((\rho_1 + 3a_1 \rho_0 - \rho_0)) \quad \dots (21)$$

and

$$a_1 = \frac{1}{2} - j \frac{k^3 a^3}{12} \left. \vphantom{\frac{1}{2} - j \frac{k^3 a^3}{12}} \right\} \text{For } a\beta \gg 1 \quad \dots (22)$$

$$P + jQ = (\rho_1 - \rho_0) / (\rho_1 + \frac{1}{2} \rho_0) \quad \dots (23)$$

The pressure due to the resultant vibration at any point situated at a distance  $x$  from the origin of disturbance is,

$$p' = 2\pi n \Delta x k a^3 [A_0 + a_1 (P + jQ)] j \omega \rho_0 \phi_0 e^{jkr} \quad \dots (24)$$

where  $\phi_0$  is the potential due to the incident waves given by,

$$\phi_0 = e^{jkr} \quad \dots (25)$$

Hence in terms of the particle velocity of the incident waves the pressure will be given by,

$$p' = 2\pi n \Delta x k a^3 [A_0 + a_1 (P + jQ)] j \omega \rho_0 u e^{jkr} \quad \dots (26)$$

where,  $u$  represents the particle velocity due to the incident waves

$$u = -jk\phi_0 \text{ or, } \phi_0 = -u/jk \quad \dots (27)$$

By putting  $x=0$  we obtain the pressure  $p'_-$  at the negative side of the stratum,

$$p'_- = 2\pi n \Delta x a^3 j \omega \rho_0 [A_0 + a_1 (P + jQ)] u \quad \dots (28)$$

Since the particles in the stratum scatter sound waves in the positive

direction as well a similar expression for  $p_+''$  is obtained,

$$p_+' = 2\pi n \Delta x a^3 j\omega \rho_0 [A_0 - a_1(P + jQ)]u. \quad \dots (29)$$

The net force per unit area of the composite medium at the ends of the stratum  $\Delta x$  acting in the negative direction will be  $(p_+' - p_-)$  i.e.,

$$p_+' - p_- = -4\pi n a^3 j\omega \rho_0 a_1 (P + jQ) \Delta x. u. \quad \dots (30)$$

### Equation of Motion

We have assumed that the particles in a gel are bound together, the exact nature of the binding forces being not known definitely. The particles give rise to scattering and the extra pressure is developed on account of this. The equation of motion is modified to,

$$\rho' \frac{du}{dt} = \frac{K'}{j\omega} \frac{d^2u}{dx^2} - 4\pi n a^3 j\omega \rho_0 a_1 (P + jQ)u \quad \dots (31)$$

or since,  $u \propto e^{j\omega t}$ , we have,

$$\frac{d^2u}{dx^2} = \frac{(j\omega)^2}{(K'/\rho')} \left[ 1 + 4\pi n a^3 \frac{\rho_0}{\rho_1} a_1 (P + jQ) \right] u \quad \dots (32)$$

putting,  $C^2 = K'/\rho'$ ,  $k = \omega/c$ , the solution of the above equation is,

$$u = e^{jk'x} \quad \dots (33)$$

where,

$$k'^2 = k^2 \left[ 1 + 4\pi n a^3 \frac{\rho_0}{\rho_1} a_1 (P + jQ) \right] \quad \dots (34)$$

or approximately, [if  $a_1(P + jQ) = p - jq$ ]

$$k' = k \left[ 1 + 2\pi n a^3 \frac{\rho_0}{\rho_1} (p - jq) \right] \quad \dots (35)$$

Hence,

$$u = e^{jk_0x} \cdot e^{-\alpha x} \quad \dots (36)$$

$$k_0 = k \left[ 1 + 2\pi n a^3 \frac{\rho_0}{\rho_1} p \right] \quad \dots (37)$$

and the attenuation constant  $\alpha$  is,

$$\alpha = 2\pi n a^3 \frac{\rho_0}{\rho_1} q.k. \quad \dots (38)$$

Thus from a knowledge of the quantities involved in the above equation the attenuation constant can be determined theoretically. Such values are given in Tables VI, VIII and IX.

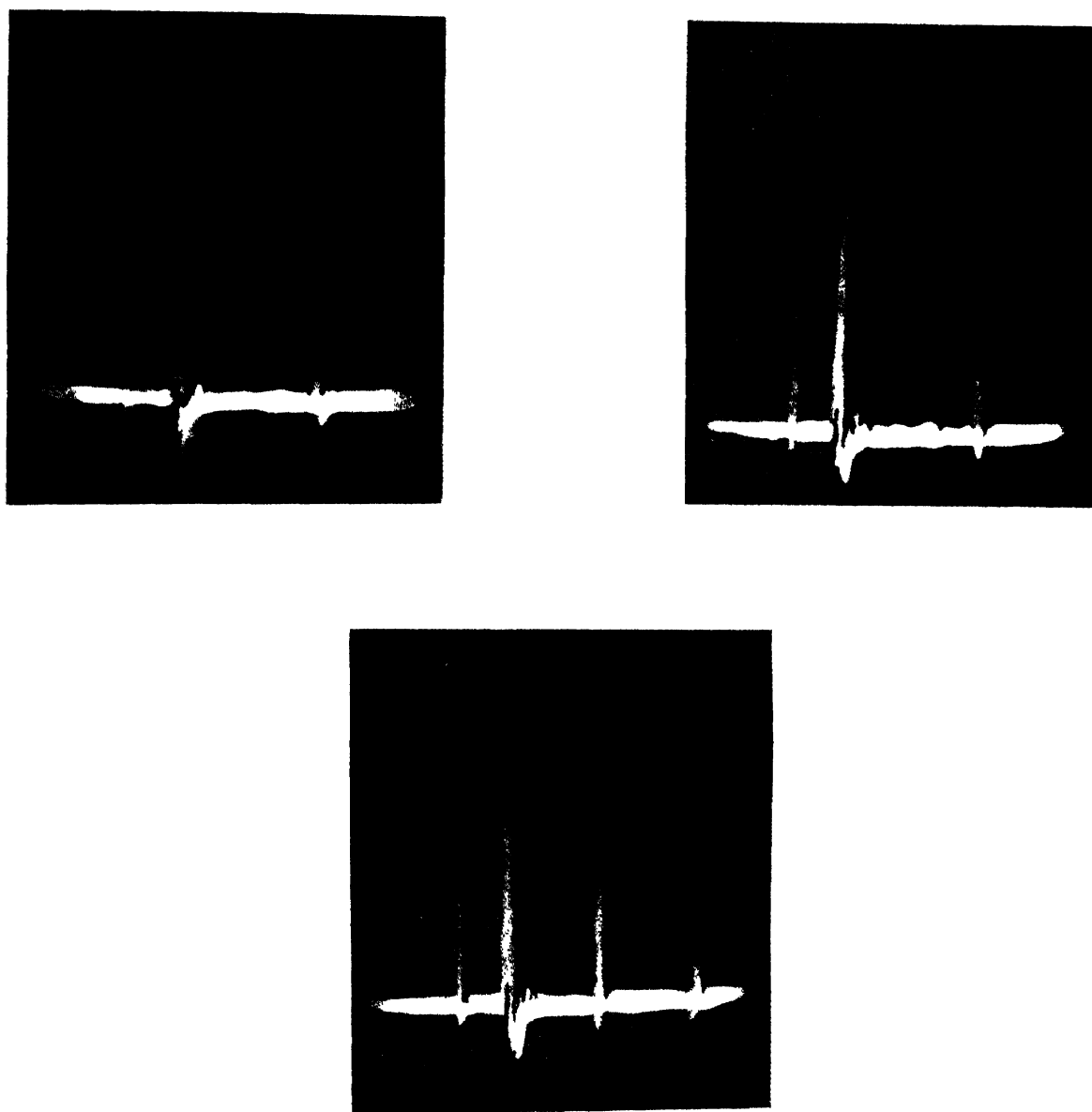
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### EXPERIMENTAL DETERMINATION OF ATTENUATION

The experimental method is based upon the successive reflection of the pulses of ultrasonic energy and an observation of the diminution in its



amplitude on an oscillograph (figure 2). The ultrasonic generator is described in detail in a previous communication (Srivastava, 1949). If



c  
FIG. 2

The oscillograms for three different values of attenuation with barium carbonate gel

$-x_1$  and  $-x_2$  are two points on the primary waves then the amplitude of the particle velocities at  $-x_1$  and  $-x_2$ , will be given by,

$$\left. \begin{aligned} u_1 &= be^{-ax_1} \\ u_2 &= be^{-ax_2} \end{aligned} \right\} \quad \dots (39)$$

and,

$$\alpha = \frac{\pi \cdot \omega}{x_1 - x_2} \log_{10} (u_1/u_2) \quad \dots (40)$$

The ratio of these yields the attenuation, since  $x_1 < x_2$ , the amplitude at  $-x_1 >$  that at  $-x_2$ . The amplitude of the particle velocity therefore decreases as the wave travels towards the negative direction of the  $x$ -axis.

The transmitter and receiver quartz are brought in contact with the two faces of the gel held in its holder. The waves are twice reflected

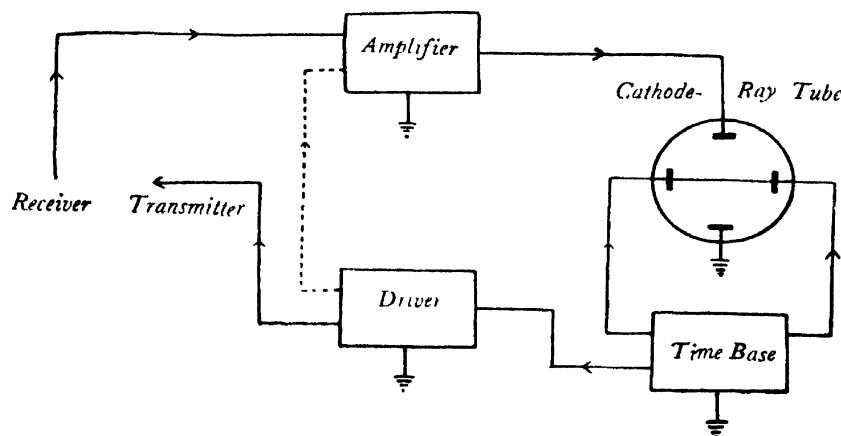


FIG. 3

The block diagram of the ultrasonic pulse generator

giving a path difference of twice the thickness of the sample, and produce two maxima on the cathode ray screen. Since reflection takes place at the surface of separation of gel and the crystal housing, a correction is applied for the loss of energy at reflection.

#### *Preparation of Gel*

The gel used in this case is barium carbonate and is prepared as follows. Fairly concentrated solutions of barium acetate and potassium carbonate are added in almost equal volumes and the mixture is allowed to stand for about 30 minutes after a vigorous shake for a minute or two. The strengths of the two solutions are determined in the usual manner, as described elsewhere (Srivastava, 1949a). In a particular case,

Strength of barium acetate, = 4.40 N.

Strength of potassium carbonate, = 4.32 N.

Density of the resulting gel obtained by mixing 5.0 c.c. each  
of the above, = 1.20 gm./cm<sup>3</sup>

An opaque stiff gel is obtained which keeps for about 3 hours. For a stiffer gel the volume of barium acetate may be greater by up to 20 per cent that of the carbonate.

RESULTS

Table VI shows a set of values of attenuation for three gels that were studied in the course of these investigations. The agreement between the theoretical and the corrected practical values is satisfactory. In Table VII the variation of attenuation due to the changes in viscosity at higher temperatures is shown. The effect of viscosity is to decrease the values of attenuation. Thus by compounding the values of Tables VI and VII it is possible to obtain the effect of both viscosity and scattering. These values are shown in Table IX. In Table VIII, only the scattering effects at higher temperatures are seen.

A detailed perusal of values in Table IX shows that the observed values are adequately explained by theoretical values. Only the results for one particular gel are given here.

TABLE VI

The values of attenuation for three gels

GEL	$\rho'$	$\beta$	$na^3$	$n$	$\alpha$ (Theory)	$\alpha$ (Corrected)	$\alpha$ (Observed)
Strontium carbonate	1.16	.052	.013	2.10	.357	.340	.418
Barium carbonate	1.20	.064	.016	2.51	.257	.306	.362
Iron silicate	1.22	.080	.020	3.10	2.48	.290	.341

TABLE VII

Changes in attenuation due to a change in viscosity at higher temperatures (eq.38).

$\nu$	$\beta$ $\times 10^5$	$1/a\beta$	$q$	$\alpha$
.010	.174	.84	1.16	.696
.008	.139	.71	.92	.552
.006	.157	.63	.78	.468
.004	.190	.52	.59	.354
.002	.240	.41	.43	.258

TABLE VIII

Changes in attenuation due to a temperature variation. Values of velocity,  $c$  have been practically determined.

Temp. °C	$\rho'$	$c$ $\times 10^5$ cms/sec	$k$	Attenuation $\alpha$		
				Practical	Corrected	Theoretical
						°C
22	1.199	2.95	25.0	.275	.306	.362 20
30	1.197	2.86	26.2	.288	.313	.370 30
43	1.192	2.73	27.5	.293	.323	.395 40
55	1.187	2.65	28.3	.311	.331	.411 50
70	1.170	2.63	28.9	.315	.300	.368 60

TABLE IX

The values of attenuation.

Temperature °C	$\rho'$	$c$ $\times 10^5$	$\beta$	Attenuation $\alpha$	
				Calculated	Observed
22	1.199	2.95	.01	.275	.306
30	1.197	2.86	.09	.288	.313
43	1.192	2.73	.008	.293	.323
55	1.187	2.65	.006	.311	.331
70	1.170	2.63	.005	.354	.300

## CONCLUSIONS

In this paper the author has tried to give a theoretical expression for the attenuation of sound waves in gelatinous substances. The structure of these substances is different from that of certain suspensions in water which have been considered by the previous workers. Upon the agreed structure of gels, which is evidenced by his work and is sufficiently supported by other distinguished sources, the author built up in this paper, considering the scattering of waves by these solid spheres, a theory that stands the test of practical experimentation. The values of attenuation are determined by an ultrasonic pulse method which has not been used elsewhere to this purpose.

Also the maxima observed in the values of attenuation have been explained as the composition of two distinct effects. The first is due to scattering which increases the value of attenuation with a rise of temperature. In the absence of any other effect this will produce a gradual increase in the attenuation. But the increase in temperature results in a decrease of kinematic viscosity, which again produces a decrease in the attenuation. Upon considering both of these, it is observed that the maximum in attenuation with temperature is suitably explained.

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